

10/031537 ✓

JC03 Rec'd PCT/PTO 18 JAN 2002

Practitioner's Docket No. YOU 2 0076**CHAPTER II**

Preliminary Classification:

Proposed Class:

Subclass:

NOTE: "All applicants are requested to include a preliminary classification on newly filed patent applications. The preliminary classification, preferably class and subclass designations, should be identified in the upper right-hand corner of the letter of transmittal accompanying the application papers, for example 'Proposed Class 2, subclass 129.'" M.P.E.P., § 601, 7th ed.

TRANSMITTAL LETTER**TO THE UNITED STATES ELECTED OFFICE (EO/US)
(ENTRY INTO U.S. NATIONAL PHASE UNDER CHAPTER II)**

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
<u>PCT/GB00/02827</u>	<u>21 July 2000 (21.07.2000)</u>	<u>30 July 1999 (30.07.1999)</u>

TITLE OF INVENTION
METHOD FOR REMOVING AND RECOVERING AROMATIC AMINES BY USING A NON-POROUS MEMBRANE

APPLICANT(S)

LIVINGSTON, Andrew Guy

Box PCT

Assistant Commissioner for Patents

Washington D.C. 20231

ATTENTION: EO/US

CERTIFICATION UNDER 37 C.F.R. §§ 1.8(a) and 1.10*
*(When using Express Mail, the Express Mail label number is mandatory;
 Express Mail certification is optional.)*

I hereby certify that, on the date shown below, this correspondence is being:

MAILING

deposited with the United States Postal Service in an envelope addressed to the Assistant Commissioner for Patents, Washington, D.C. 20231

37 C.F.R. § 1.8(a)37 C.F.R. § 1.10 *

with sufficient postage as first class mail as "Express Mail Post Office to Addressee"
 Mailing Label No ET 852686949 US (mandatory)

TRANSMISSION

facsimile transmitted to the Patent and Trademark Office, (703) _____

Date: 11/18/02

Signature

Georange B. GeorgeGeorange B. George

(type or print name of person certifying)

* Only the date of filing (§ 1.6) will be the date used in a patent term adjustment calculation, although the date on any certificate of mailing or transmission under § 1.8 continues to be taken into account in determining timeliness. See § 1.703(f). Consider "Express Mail Post Office to Addressee" (§ 1.10) or facsimile transmission (§ 1.6(d)) for the reply to be accorded the earliest possible filing date for patent term adjustment calculations.

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NOTE: To avoid abandonment of the application, the applicant shall furnish to the USPTO, not later than 20 months from the priority date: (1) a copy of the international application, unless it has been previously communicated by the International Bureau or unless it was originally filed in the USPTO; and (2) the basic national fee (see 37 C.F.R. § 1.492(a)). The 30-month time limit may not be extended. 37 C.F.R. § 1.495.

WARNING: Where the items are those which can be submitted to complete the entry of the international application into the national phase are subsequent to 30 months from the priority date the application is still considered to be in the international state and if mailing procedures are utilized to obtain a date the express mail procedure of 37 C.F.R. § 1.10 must be used (since international application papers are not covered by an ordinary certificate of mailing—See 37 C.F.R. § 1.8).

NOTE: Documents and fees must be clearly identified as a submission to enter the national state under 35 U.S.C. § 371 otherwise the submission will be considered as being made under 35 U.S.C. § 111. 37 C.F.R. § 1.494(f).

I. Applicant herewith submits to the United States Elected Office (EO/US) the following items under 35 U.S.C. § 371:

- a. This express request to immediately begin national examination procedures (35 U.S.C. § 371(f)).
- b. The U.S. National Fee (35 U.S.C. § 371(c)(1)) and other fees (37 C.F.R. § 1.492) as indicated below:

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2. Fees

CLAIMS FEE	(1) FOR	(2) NUMBER FILED	(3) NUMBER EXTRA	(4) RATE	(5) CALCULA- TIONS
XX*	TOTAL CLAIMS	32 - 20 =	12	× \$18.00 =	\$ 216.00
	INDEPENDENT CLAIMS	1 - 3 =	0	× \$84.00 =	0
	MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$280.00	0
BASIC FEE**	<input type="checkbox"/> U.S. PTO WAS INTERNATIONAL PRELIMINARY EXAMINATION AUTHORITY Where an International preliminary examination fee as set forth in § 1.482 has been paid on the international application to the U.S. PTO: <input type="checkbox"/> and the international preliminary examination report states that the criteria of novelty, inventive step (non-obviousness) and industrial activity, as defined in PCT Article 33(1) to (4) have been satisfied for all the claims presented in the application entering the national stage (37 C.F.R. § 1.492(a)(4)) \$100.00 <input type="checkbox"/> and the above requirements are not met (37 C.F.R. § 1.492(a)(1)) \$710.00				
	<input checked="" type="checkbox"/> U.S. PTO WAS NOT INTERNATIONAL PRELIMINARY EXAMINATION AUTHORITY Where no international preliminary examination fee as set forth in § 1.482 has been paid to the U.S. PTO, and payment of an international search fee as set forth in § 1.445(a)(2) to the U.S. PTO: <input type="checkbox"/> has been paid (37 C.F.R. § 1.492(a)(2)) \$740.00 <input type="checkbox"/> has not been paid (37 C.F.R. § 1.492(a)(3)) \$1040.00 <input checked="" type="checkbox"/> where a search report on the international application has been prepared by the European Patent Office or the Japanese Patent Office (37 C.F.R. § 1.492(a)(5)) \$890.00				890.00
		Total of above Calculations	=	1,146.00	
SMALL ENTITY	Reduction by 1/2 for filing by small entity, if applicable. Assertion must be made. (note 37 C.F.R. § 1.27)			-	
		Subtotal		1,106.00	
		Total National Fee	\$	1,106.00	
	Fee for recording the enclosed assignment document \$40.00 (37 C.F.R. § 1.21(h)). (See Item 13 below). See attached "ASSIGNMENT COVER SHEET".			40.00	
TOTAL		Total Fees enclosed	\$	1,146.00	

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*See attached Preliminary Amendment Reducing the Number of Claims.

- Attached is a check money order in the amount of \$ 1,146.00
- Authorization is hereby made to charge the amount of \$ 0
 - to Deposit Account No. 06-0308
 - to Credit card as shown on the attached credit card information authorization form PTO-2038.

WARNING: Credit card information should not be included on this form as it may become public.

- Charge any additional fees required by this paper or credit any overpayment in the manner authorized above.

A duplicate of this paper is attached.

WARNING: "To avoid abandonment of the application the applicant shall furnish to the United States Patent and Trademark Office not later than the expiration of 30 months from the priority date: * * * (2) the basic national fee (see § 1.492(a)). The 30-month time limit may not be extended." 37 C.F.R. § 1.495(b).

WARNING: If the translation of the international application and/or the oath or declaration have not been submitted by the applicant within thirty (30) months from the priority date, such requirements may be met within a time period set by the Office. 37 C.F.R. § 1.495(b)(2). The payment of the surcharge set forth in § 1.492(e) is required as a condition for accepting the oath or declaration later than thirty (30) months after the priority date. The payment of the processing fee set forth in § 1.492(f) is required for acceptance of an English translation later than thirty (30) months after the priority date. Failure to comply with these requirements will result in abandonment of the application. The provisions of § 1.136 apply to the period which is set. Notice of Jan. 3, 1993, 1147 O.G. 29 to 40.

- Assertion of Small Entity Status
- Applicant hereby asserts status as a small entity under 37 C.F.R. § 1.27.

NOTE: 37 C.F.R. § 1.27(c) deals with the assertion of small entity status, whether by a written specific declaration thereof or by payment as a small entity of the basic filing fee or the fee for the entry into the national phase as states:

"(c) Assertion of small entity status. Any party (person, small business concern or nonprofit organization) should make a determination, pursuant to paragraph (f) of this section, of entitlement to be accorded small entity status based on the definitions set forth in paragraph (a) of this section, and must, in order to establish small entity status for the purpose of paying small entity fees, actually make an assertion of entitlement to small entity status, in the manner set forth in paragraphs (c)(1) or (c)(3) of this section, in the application or patent in which such small entity fees are to be paid.

(1) Assertion by writing. Small entity status may be established by a written assertion of entitlement to small entity status. A written assertion must:

- (i) Be clearly identifiable;
- (ii) Be signed (see paragraph (c)(2) of this section); and
- (iii) Convey the concept of entitlement to small entity status, such as by stating that applicant is a small entity, or that small entity status is entitled to be asserted for the application or patent. While no specific words or wording are required to assert small entity status, the intent to assert small entity status must be clearly indicated in order to comply with the assertion requirement.

(2) Parties who can sign and file the written assertion. The written assertion can be signed by:

- (i) One of the parties identified in §§ 1.33(b) (e.g., an attorney or agent registered with the Office), §§ 3.73(b) of this chapter notwithstanding, who can also file the written assertion;
- (ii) At least one of the individuals identified as an inventor (even though a §§ 1.63 executed oath or declaration has not been submitted), notwithstanding §§ 1.33(b)(4), who can also file the written assertion pursuant to the exception under §§ 1.33(b) of this part; or
- (iii) An assignee of an undivided part interest, notwithstanding §§ 1.33(b)(3) and 3.73(b) of this chapter, but the partial assignee cannot file the assertion without resort to a party identified under §§ 1.33(b) of this part.

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(3) Assertion by payment of the small entity basic filing or basic national fee. The payment, by any party, of the exact amount of one of the small entity basic filing fees set forth in §§ 1.16(e), (f), (g), (h), or (k), or one of the small entity basic national fees set forth in §§ 1.492(a)(1), (a)(2), (a)(3), (a)(4), or (a)(5), will be treated as a written assertion of entitlement to small entity status even if the type of basic filing or basic national fee is inadvertently selected in error.

(i) If the Office accords small entity status based on payment of a small entity basic filing or basic national fee under paragraph (c)(3) of this section that is not applicable to that application, any balance of the small entity fee that is applicable to that application will be due along with the appropriate surcharge set forth in §§ 1.16(e), or §§ 1.16(f).

(ii) The payment of any small entity fee other than those set forth in paragraph (c)(3) of this section (whether in the exact fee amount or not) will not be treated as a written assertion of entitlement to small entity status and will not be sufficient to establish small entity status in an application or a patent.*

3. A copy of the International application as filed (35 U.S.C. § 371(c)(2)).

NOTE: Section 1.495 (b) was amended to require that the basic national fee and a copy of the international application must be filed with the Office by 30 months from the priority date to avoid abandonment. "The International Bureau normally provides the copy of the International application to the Office in accordance with PCT Article 20. At the same time, the International Bureau notifies applicant of the communication to the Office. In accordance with PCT Rule 47.1, that notice shall be accepted by all designated offices as conclusive evidence that the communication has duly taken place. Thus, if the applicant desires to enter the national stage, the applicant normally need only check to be sure the notice from the International Bureau has been received and then pay the basic national fee by 30 months from the priority date." Notice of Jan. 7, 1993, 1147 O.G. 29 to 40, at 35-36. See item 14c below.

- a. is transmitted herewith.
- b. is not required, as the application was filed with the United States Receiving Office.
- c. has been transmitted
 - i. by the International Bureau.

Date of mailing of the application (from form PCT/1B/308): _____

ii. by applicant on _____. (Date)

4. A translation of the International application into the English language (35 U.S.C. § 371(c)(2)).

- a. is transmitted herewith.
- b. is not required as the application was filed in English.
- c. was previously transmitted by applicant on _____. (Date)
- d. will follow.

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5. Amendments to the claims of the International application under PCT Article 19 (35 U.S.C. § 371(c)(3));

NOTE: The Notice of January 7, 1993 points out that 37 C.F.R. § 1.495(a) was amended to clarify the existing and continuing practice that PCT Article 19 amendments must be submitted by 30 months from the priority date and this deadline may not be extended. The Notice further advises that: "The failure to do so will not result in loss of the subject matter of the PCT Article 19 amendments. Applicant may submit that subject matter in a preliminary amendment filed under section 1.121. In many cases, filing an amendment under section 1.121 is preferable since grammatical or idiomatic errors may be corrected." 1147 O.G. 29-40, at 36.

- a. are transmitted herewith.
- b. have been transmitted
 - i. by the International Bureau.

Date of mailing of the amendment (from form PCT/1B/308):

- ii. by applicant on _____, (Date)
- c. have not been transmitted as
 - i. applicant chose not to make amendments under PCT Article 19. Date of mailing of Search Report (from form PCT/ISA/210.); December 19, 2000.
 - ii. the time limit for the submission of amendments has not yet expired. The amendments or a statement that amendments have not been made will be transmitted before the expiration of the time limit under PCT Rule 46.1.

6. A translation of the amendments to the claims under PCT Article 19 (38 U.S.C. § 371(c)(3));
 - a. is transmitted herewith.
 - b. is not required as the amendments were made in the English language.
 - c. has not been transmitted for reasons indicated at point 5(c) above.
7. A copy of the international examination report (PCT/IPEA/409)
 - is transmitted herewith.
 - is not required as the application was filed with the United States Receiving Office.
8. Annex(es) to the International preliminary examination report
 - a. is/are transmitted herewith.
 - b. is/are not required as the application was filed with the United States Receiving Office.
9. A translation of the annexes to the international preliminary examination report
 - a. is transmitted herewith.
 - b. is not required as the annexes are in the English language.

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10. An oath or declaration of the inventor (35 U.S.C. § 371(c)(4)) complying with 35 U.S.C. § 115

- a. was previously submitted by applicant on _____ (Date)
- b. is submitted herewith, and such oath or declaration
 - i. is attached to the application.
 - ii. Identifies the application and any amendments under PCT Article 19 that were transmitted as stated in points 3(b) or 3(c) and 5(b); and states that they were reviewed by the inventor as required by 37 C.F.R. § 1.70.
- c. will follow.

II. Other document(s) or information included:

11. An International Search Report (PCT/ISA/210) or Declaration under PCT Article 17(2)(e):

- a. is transmitted herewith.
- b. has been transmitted by the International Bureau.
Date of mailing (from form PCT/IB/308): _____
- c. is not required, as the application was searched by the United States International Searching Authority.
- d. will be transmitted promptly upon request.
- e. has been submitted by applicant on _____. (Date)

12. An Information Disclosure Statement under 37 C.F.R. §§ 1.97 and 1.98:

- a. is transmitted herewith.

Also transmitted herewith is/are:

- Form PTO-1449 (PTO/SB/08A and 08B).
- Copies of citations listed.
- b. will be transmitted within THREE MONTHS of the date of submission of requirements under 35 U.S.C. § 371(c).
- c. was previously submitted by applicant on _____. (Date)

13. An assignment document is transmitted herewith for recording.

A separate "COVER SHEET FOR ASSIGNMENT (DOCUMENT) ACCOMPANYING NEW PATENT APPLICATION" or FORM PTO 1595 is also attached.

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531 Receipt/PCTC 12 JAN 2002

14. Additional documents:

- a. Copy of request (PCT/RO/101)
- b. International Publication No. WO 01/09042 A1
 - i. Specification, claims and drawing
 - ii. Front page only
- c. Preliminary amendment (37 C.F.R. § 1.121)
- d. Other
PCT/ISA/220

15. The above checked items are being transmitted

- a. before 30 months from any claimed priority date.
- b. after 30 months.

16. Certain requirements under 35 U.S.C. § 371 were previously submitted by the applicant on _____, namely:

AUTHORIZATION TO CHARGE ADDITIONAL FEES

WARNING: Accurately count claims, especially multiple defendant claims, to avoid unexpected high charges if extra claims are authorized.

NOTE: "A written request may be submitted in an application that is an authorization to treat any concurrent or future reply, requiring a petition for an extension of time under this paragraph for its timely submission, as incorporating a petition for extension of time for the appropriate length of time. An authorization to charge all required fees, fees under § 1.17, or all required extension of time fees will be treated as a constructive petition for an extension of time in any concurrent or future reply requiring a petition for an extension of time under this paragraph for its timely submission. Submission of the fee set forth in § 1.17(a) will also be treated as a constructive petition for an extension of time in any concurrent reply requiring a petition for an extension of time under this paragraph for its timely submission." 37 C.F.R. § 1.136(a)(3).

NOTE: "Amounts of twenty-five dollars or less will not be returned unless specifically requested within a reasonable time, nor will the payer be notified of such amounts; amounts over twenty-five dollars may be returned by check or, if requested, by credit to a deposit account." 37 C.F.R. § 1.26(a).

Please charge, in the manner authorized above, the following additional fees that may be required by this paper and during the entire pendency of this application:

37 C.F.R. § 1.492(a)(1), (2), (3), and (4) (filing fees)

WARNING: Because failure to pay the national fee within 30 months without extension (37 C.F.R. § 1.495(b)(2)) results in abandonment of the application, it would be best to always check the above box.

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37 C.F.R. § 1.492(b), (c) and (d) (presentation of extra claims)

NOTE: Because additional fees for excess or multiple dependent claims not paid on filing or on later presentation must only be paid or these claims cancelled by amendment prior to the expiration of the time period set for response by the PTO in any notice of fee deficiency (37 C.F.R. § 1.492(d)), it might be best not to authorize the PTO to charge additional claim fees, except possible when dealing with amendments after final action.

37 C.F.R. § 1.17 (application processing fees)
 37 C.F.R. § 1.17(a)(1)-(5) (extension fees pursuant to § 1.136(a)).
 37 C.F.R. § 1.18 (issue fee at or before mailing of Notice of Allowance, pursuant to 37 C.F.R. § 1.311(b))

NOTE: Where an authorization to charge the issue fee to a deposit account has been filed before the mailing of a Notice of Allowance, the issue fee will be automatically charged to the deposit account at the time of mailing the notice of allowance. 37 C.F.R. § 1.311(b).

NOTE: 37 C.F.R. § 1.28(b) requires "Notification of any change in loss of entitlement to small entity status must be filed in the application . . . prior to paying, or at the time of paying . . . issue fee." From the wording of 37 C.F.R. § 1.28(b): (a) notification of change of status must be made even if the fee is paid as "other than a small entity" and (b) no notification is required if the change is to another small entity.

37 C.F.R. § 1.492(e) and (f) (surcharge fees for filing the declaration and/or filing an English translation of an International Application later than 30 months after the priority date).



SIGNATURE OF PRACTITIONER

Scott A. McCollister

(type or print name of practitioner)

Fay, Sharpe, Fagan, Minnich & McKee, LLP

P.O. Address

1100 Superior Avenue, Seventh Floor
Cleveland, Ohio 44114-2518

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Tel. No.: (216) 861-5582

Customer No.:

10/031537
531 Rec'd PCT/FTC 18 JAN 2002

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF : LIVINGSTON, Andrew Guy
FOR : METHOD FOR REMOVING AND
RECOVERING AROMATIC AMINES
BY USING A NON-POROUS
MEMBRANE
SERIAL NO. : Unknown
FILED : Herewith
ATTORNEY DOCKET NO. : YOU 2 0076

Cleveland, Ohio 44114-2518
January 18, 2002

PRELIMINARY AMENDMENT

Assistant Commissioner For Patents
Washington, D.C. 20231

Dear Sir:

Prior to calculation of the filing fee and substantive examination of the above-referenced patent application, Applicants respectfully request amendment of the application as follows. A clean copy of the claims appears below and a marked-up version is attached as an appendix.

IN THE CLAIMS:

Please delete claim 33.

Please amend the claims as follows:

3. (Amended) A process according to claim 1 wherein the aqueous fluid is contacted with one side of the membrane and wherein the acidic stripping solution is contacted with the other side of the membrane.

5. (Amended) A process according to claim 3 wherein the acidic stripping solution separated in step (e) is recycled to the aqueous fluid prior to contact with the membrane.
7. (Amended) A process according to claim 1 wherein the resulting aromatic amine of step (e) is a liquid.
8. (Amended) A process according to claim 1 wherein the resulting aromatic amine of step (e) is a solid.
9. (Amended) A process according to claim 1 wherein the membrane is mounted in a plate and frame configuration, a shell and tube configuration, or a spiral wound configuration.
10. (Amended) A process according to claim 1 wherein the membrane comprises one or more tubular membranes each of which has an internal surface which defines an internal volume and either the aqueous fluid or the acidic stripping solution is held within the internal volume of the tubular membrane(s) and the other of the aqueous fluid or the acidic stripping solution is in contact with the external surface of the tubular membrane(s).
12. (Amended) A process according to claim 10 wherein the tubular membrane(s) is elastomeric.
13. (Amended) A process according to claim 10 wherein the tubular membrane(s) has a length to diameter ratio of from 10,000 to 1,000,000.
14. (Amended) A process according to claim 1 wherein the acidic stripping solution in contact with the nonporous membrane is well mixed throughout its volume such that its composition is uniform throughout.

15. (Amended) A process according to claim 1 wherein the pH of the stripping solution in contact with the non-porous membrane is controlled so that it is substantially the same throughout the volume of acidic stripping solution in contact with the non-porous membrane separating layer.

16. (Amended) A process according to claim 1 wherein the aqueous fluid contains an aromatic amine selected from aniline, chloroanilines, dichloroanilines, toluidines, dimethylaniline, nitroaniline, phenylaniline, methylaniline, ethylaniline, anisidine, benzylamine, fluoroanilines, difluoroanilines, aminophenols and mixtures thereof.

17. (Amended) A process according to claim 1 wherein the acidic stripping solution comprises a mineral acid selected from hydrochloric acid, sulphuric acid, phosphoric acid, nitric acid, and mixtures thereof.

18. (Amended) A process according to claim 1 wherein the pH of the aromatic amine containing acidic stripping solution is adjusted in step (d) by the addition of an alkali.

20. (Amended) A process according to claim 1 wherein the membrane is formed from or comprises a material selected from modified polysiloxane based elastomers, ethylene-propylene diene (EPDM) based elastomers, polynorbornene based elastomers, polyoctenamer based elastomers, polyurethane based elastomers, butadiene and nitrile butadiene rubber based elastomers, natural rubber, butyl rubber based elastomers, polychloroprene (Neoprene) based elastomers, epichlorohydrin elastomers, polyacrylate elastomers, polyethylene, polypropylene, polytetrafluoroethylene (PTFE), polyvinylidene difluoride (PVDF) based elastomers, and mixtures thereof.

22. (Amended) A process according to claim 1 wherein the membrane comprises a reinforcing material selected from an external mesh and support.

23. (Amended) A process according to claim 1 wherein the membrane is a composite membrane comprising a porous support and at least one non-porous layer.

25. (Amended) A process according to claim 1 wherein the aqueous fluid is contacted with one side of a plurality of membranes in series, in parallel or in a combination thereof, and wherein the acidic stripping solution is contacted with the other side of each of the plurality of membranes.

26. (Amended) A process according to claim 1 wherein the process is performed in a continuous manner.

27. (Amended) A process according to claim 19 wherein the flow of at least one of the aqueous fluid, the acidic stripping solution, and the alkali solution is discontinuous.

28. (Amended) A process according to claim 1 wherein the resulting aromatic amine of step (e) is contacted with an organic solvent and subsequently treated in a further process.

29. (Amended) A process according to claim 1 wherein the process is performed in a reactor comprising at least a first zone, a second zone, a third zone, and a fourth zone;
wherein each of the zones is discrete from each other zone;
wherein the first zone and the second zone are separated by the non-porous membrane;
wherein the first zone contains the aqueous fluid;
wherein the second zone and fourth zone contain the acidic stripping solution;
wherein the third zone contains amine containing acidic stripping solution;
wherein the first zone and the third zone are operably connected to each other;

wherein the second zone is operably connected to the third zone and the fourth zone; and

wherein the acidic stripping solution is circulated between the fourth zone and the second zone such that the acidic stripping solution is well mixed throughout its volume.

31. (Amended) A process according to claim 1 wherein at least one of the aqueous fluid and the acidic stripping solution has a temperature above room temperature.

32. (Amended) A process according to claim 1 wherein the aqueous fluid contains substantial quantities of dissolved inorganic or organic materials.

Remarks

Applicants respectfully request that the foregoing amendments be entered prior to calculation of the filing fee and substantive examination of the application.

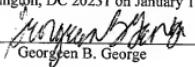
Respectfully submitted,
FAY, SHARPE, FAGAN
MINNICH & McKEE, LLP

Date: January 18, 2002


Scott A. McCollister
Reg. No. 33,961
1100 Superior Avenue, 7th Floor
Cleveland, Ohio 44114-2518
(216) 861-5582

CERTIFICATE OF MAILING

I hereby certify that this PRELIMINARY AMENDMENT is being deposited with the United States Postal Service as EXPRESS MAIL in an envelope numbered EL805293170 US, addressed to Assistant Commissioner for Patents, Washington, DC 20231 on January 18, 2002.

By: 
Georgene B. George

VERSION WITH MARKINGS TO SHOW CHANGES MADE

3. (Amended) A process according to claim 1 [or 2] wherein the aqueous fluid is contacted with one side of the membrane and wherein the acidic stripping solution is contacted with the other side of the membrane.

5. (Amended) A process according to claim 3 [or 4] wherein the acidic stripping solution separated in step (e) is recycled to the aqueous fluid prior to contact with the membrane.

7. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the resulting aromatic amine of step (e) is a liquid.

8. (Amended) A process according to [any one of claims 1 to 6] claim 1 wherein the resulting aromatic amine of step (e) is a solid.

9. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the membrane is mounted in a plate and frame configuration, a shell and tube configuration, or a spiral wound configuration.

10. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the membrane comprises one or more tubular membranes each of which has an internal surface which defines an internal volume and either the aqueous fluid or the acidic stripping solution is held within the internal volume of the tubular membrane(s) and the other of the aqueous fluid or the acidic stripping solution is in contact with the external surface of the tubular membrane(s).

12. (Amended) A process according to claim 10 [or 11] wherein the tubular membrane(s) is elastomeric.

13. (Amended) A process according to [any one of claims 10 to 12] claim 10 wherein the tubular membrane(s) has a length to diameter ratio of from 10,000 to 1,000,000.

14. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the acidic stripping solution in contact with the nonporous membrane is well mixed throughout its volume such that its composition is uniform throughout.

15. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the pH of the stripping solution in contact with the non-porous membrane is controlled so that it is substantially the same throughout the volume of acidic stripping solution in contact with the non-porous membrane separating layer.

16. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the aqueous fluid contains an aromatic amine selected from aniline, chloroanilines, dichloroanilines, toluidines, dimethylaniline, nitroaniline, phenylaniline, methylaniline, ethylaniline, anisidine, benzylamine, fluoroanilines, difluoroanilines, aminophenols and mixtures thereof.

17. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the acidic stripping solution comprises a mineral acid selected from hydrochloric acid, sulphuric acid, phosphoric acid, nitric acid, and mixtures thereof

18. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the pH of the aromatic amine containing acidic stripping solution is adjusted in step (d) by the addition of an alkali.

20. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the membrane is formed from or comprises a material selected from modified polysiloxane based elastomers, ethylene-propylene diene (EPDM) based elastomers, polynorbornene based elastomers, polyoctenamer based elastomers, polyurethane based elastomers, butadiene and nitrile butadiene rubber based elastomers, natural rubber, butyl rubber based elastomers, polychloroprene (Neoprene) based elastomers, epichlorohydrin elastomers, polyacrylate elastomers, polyethylene, polypropylene, polytetrafluoroethylene (PTFE), polyvinylidene difluoride (PVDF) based elastomers, and mixtures thereof.

22. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the membrane comprises a reinforcing material selected from an external mesh and support.

23. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the membrane is a composite membrane comprising a porous support and at least one non-porous layer.

25. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the aqueous fluid is contacted with one side of a plurality of membranes in series, in parallel or in a combination thereof, and wherein the acidic stripping solution is contacted with the other side of each of the plurality of membranes.

26. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the process is performed in a continuous manner.

27. (Amended) A process according to [any one of claims 19 to 25] claim 19 wherein the flow of at least one of the aqueous fluid, the acidic stripping solution, and the alkali solution is discontinuous.

28. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the resulting aromatic amine of step (e) is contacted with an organic solvent and subsequently treated in a further process.

29. (Amended) A process according to [any one of the preceding claims] claim 1 wherein the process is performed in a reactor comprising at least a first zone, a second zone, a third zone, and a fourth zone; wherein each of the zones is discrete from each other zone; wherein the first zone and the second zone are separated by the non-porous membrane; wherein the first zone contains the aqueous fluid; wherein the second zone and fourth zone contain the acidic stripping solution; wherein the third zone contains amine containing acidic stripping solution; wherein the first zone and the third zone are operably connected to each other; wherein the second zone is operably connected to the third zone and the fourth zone; and wherein the acidic stripping solution is circulated between the fourth zone and the second zone such that the acidic stripping solution is well mixed throughout its volume.

31. (Amended) A process according to [any one of the preceding claims] claim 1 wherein at least one of the aqueous fluid and the acidic stripping solution has a temperature above room temperature.

32. (Amended) A process according to [any of the preceding claims] claim 1 wherein the aqueous fluid contains substantial quantities of dissolved inorganic or organic materials.

**METHOD FOR REMOVING AND RECOVERING AROMATIC AMINES BY
USING A NON-POROUS MEMBRANE**

The present invention relates to a process for the removal and recovery of one or more aromatic amines from an aqueous fluid. In particular the process comprises transferring 5 the aromatic amines from the aqueous fluid to an acidic stripping solution across a non porous, selectively permeable membrane, adjusting the pH of the acidic stripping solution and separating the resulting aromatic amine from the acidic stripping solution.

Many organic bases, such as aniline, toluidine, nitroaniline, enter aqueous process streams 10 in chemical processing. These molecules are in many cases toxic. Methods for removing toxic organic molecules from aqueous process streams are well known. Some of these methods use membranes.

Membrane solvent extraction using microporous membranes to provide a phase 15 contacting between aqueous and organic streams is well known. For example Kiani, Bhave and Sirkar Journal of Membrane Science 20 (1984) pp 125-145 report the use of microporous membranes for immobilising solvent interfaces during solvent extraction. Tompkins, Micheals and Peretti Journal of Membrane Science 75 (1992) pp 277-292 report using microporous polypropylene fibres to stabilise phase interfaces during 20 extraction of nitrophenol from an aqueous solution into octanol. US 5,512,180 describes a process wherein polypropylene glycol MW 4000 was used to extract nitrophenol in a microporous membrane contactor.

A continuing problem with membrane supported solvent extraction with microporous 25 membranes is the breakthrough of one phase into the other due to pressure imbalances. To overcome this problem, various improvements have been suggested such as using composite membranes comprising a thin layer of non-porous organic-permeable polymer bound to a microporous membrane to avoid phase breakthrough, for example US 4,960,520. However, in all of these processes a solvent phase containing the organic 30 compound is produced which must then be disposed of or treated in some way.

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Contacting two aqueous streams with opposite sides of a membrane to effect extraction of organic pollutants from one side to the other is also known in the art. Supported liquid membranes have been applied in this mode. For example US 5,507,949 describes a process wherein the pores of a microporous hydrophobic membrane are filled with a hydrophobic polyamphiphilic oligomeric or polymeric liquid to allow mass transport of various organics across the membranes. In this application the driving force for extraction across the supported liquid membranes may be provided by a stripping solution. The driving force produced by a stripping solution may rely upon conversion of an organic acid to its corresponding salt using a basic stripping solution, or conversion of an organic base to its corresponding salt using an acidic stripping solution. Biologically active stripping solutions may also be utilised, for example US 4,988,443 to Michaels et al. discloses a method for contacting an aqueous waste stream containing organic toxicants with a nutrient-containing aqueous stream using hollow fibre membranes with water immiscible solvent filled pores. The two streams do not mix but the organic toxicants are transferred from the waste stream across the membrane to the nutrient stream. Microorganisms growing associated with the outside of the hollow fibres utilise the nutrients and organic toxicants as growth substrates which provides the driving force for continued transport.

In further applications non-porous membranes have been employed to effect extraction of organic molecules from one aqueous stream into another. US 5,552,053 discloses solid polyamphiphilic polymer films used for keeping separate two aqueous phases, one being a waste stream and the other a stripping solution in which the organic pollutant can be concentrated by conversion into an ionised form at controlled pH.

In the above prior art, membranes are substantially rigid and are employed in shell and tube modules, in plate and frame modules, or in spiral wound modules. These modules are designed to generate good mass transfer and fluid distribution around all of the membrane surfaces.

In a few cases, tubular elastomeric non-porous homogeneous membranes for example

silicone rubber (cross linked polydimethoxysiloxane) tubes have been disclosed. The tubular elastomeric membranes provide separation by allowing specific chemical species (for example, hydrophobic organic molecules such as benzene, toluene, or their derivatives) to preferentially dissolve in the membrane and permeate across the membrane
5 by diffusion under the influence of a chemical activity driving force. For example, US 5,585,004 to Livingston discloses a system of apparatus and method wherein a waste stream containing toxic organic compounds is fed to the inside of selectively permeable silicone rubber membrane tubes suspended in a bioreactor receptacle filled with a biologically active medium. The toxic organic compounds diffuse across the silicone
10 rubber membrane and into the biologically active medium where they are destroyed by the microbial culture.

Further examples of the use of tubular elastomeric membranes are oxygenation of microbial systems (Cote et al, Journal of Membrane Science 1989 47 p107), and pervaporation (Raghunath and Hwang, Journal of Membrane Science 1992 65 p147). In the field of chemical analysis, silicone rubber membranes have been used to extract organics from aqueous streams prior to analysis (US 4,715,217; US 4,891,137).

The processing of organic-laden stripping solutions comprising organic acids in
20 dissociated form in an aqueous solution is known with regard to nitrophenolic compounds recovery. For example, various processes are known in the art for disposing of stripping solutions containing nitrophenolic materials. These stripping solutions are generated as a by-product of nitration reactions. US Patent 4,597,875 discloses a process for removing the nitrophenolic materials from an alkaline stripping solution by treating the wastewater
25 with an acid to lower its pH and convert the nitrophenolic compounds to a water insoluble solid material which is separated out of the wastewater and can be disposed of by incineration. US 4,925,565 discusses a process in which the alkaline stripping solution is treated with acid to lower its pH, following which a substantially water insoluble solvent is used to extract the nitrophenolic compounds from the wastewater at elevated
30 temperature. The solvent is recovered by distillation and the residue containing nitrophenolics can be incinerated. In variations on US 4,925,565, the same inventors use

differential control of the pH to recover specific nitrophenolic fractions by solvent extraction (US 4,986,917) and precipitation (US 4,986,920). However, the recovery of the nitrophenolic fraction is complicated by the fact that the nitrophenols form solid precipitates upon acidification of alkaline wastewaters containing ionised nitrophenolic compounds at concentrations above the saturation concentration of non-ionised nitrophenolic compounds in water.

In the prior art utilising membranes for organics removal, the focus has been on the use of alkaline stripping solutions to remove organic acids from aqueous fluids. Many polymeric membrane materials and in particular elastomers have good resistance to alkalis. However, when it is desired to remove organic bases such as aromatic amines using acidic stripping solutions, problems can arise due to the lack of chemical resistance of many polymeric membrane materials, in particular elastomers, to mineral acids.

15 The present invention addresses the problems of the prior art.

In one aspect the present invention provides a process for removing and recovering one or more undissociated aromatic amines dissolved in aqueous fluid, the process comprising the steps of: (a) transferring the one or more undissociated aromatic amines from the aqueous fluid to an acidic stripping solution, wherein transfer of the one or more undissociated aromatic amines from the aqueous fluid to the acidic stripping solution occurs across a membrane; wherein the membrane is a non porous, selectively permeable membrane; (b) regulating the volume of acidic stripping solution employed relative to the volume of aqueous fluid treated so that the total aromatic amine concentration in the acidic stripping solution, comprising the sum of the dissociated and undissociated aromatic amine concentrations, is above the solubility of the aromatic amines in water; (c) regulating the pH of the acidic stripping solution in contact with the membrane so that the membrane remains selectively permeable; (d) adjusting the pH of the aromatic amine containing acidic stripping solution to a value above the acidic dissociation constant of the aromatic amine and (e) separating the resulting aromatic amine and the acidic stripping solution.

By the term "selectively permeable" it is meant a membrane which is permeable to the undissociated aromatic amine and which is impermeable to the dissociated aromatic amine.

5

It will be appreciated that the term "aromatic amine" includes any compound of the formula $R^1NR^2R^3$ wherein R^1 is or comprises an aromatic group and R^2 and R^3 are independently selected from H or a hydrocarbyl group, such an alkyl or an aryl group. In this aspect an undissociated aromatic amine is a compound of the formula $R^1NR^2R^3$ and a dissociated aromatic amine is a compound of the formula $R^1NR^2R^3H^+$.

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Step (c) requires that the pH of the acidic stripping solution in contact with the membrane is regulated so that the membrane remains selectively permeable. By this it is meant that the pH should be regulated so that the membrane materials are not rendered unusable through attack by the acidic stripping solution.

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The present inventors have found that control of the pH in the acidic stripping solution assists in the reducing of costs and in increasing the membrane lifetime, particularly in respect of acidic stripping solutions.

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In the present invention, aromatic amines present in an aqueous fluid in undissociated form are recovered by means of membrane extraction across a membrane. The membrane contains at least one non porous, selectively permeable layer. The aromatic amines pass into an acidic stripping solution in which the aromatic amines undergo dissociation. The acidic stripping solution is then further processed by adjusting the pH upwards until the aromatic amines re-associate and precipitate out of solution as liquids or solids.

25

Aromatic amines will undergo a dissociation reaction when the pH of the stripping solution is below the $pK_a = (14-pK_b)$ of the aromatic amine, where pK_b is the basicity constant and is defined as follows (see for example "Organic Chemistry" third Edition by T.W.G.Solomons. John Wiley and Sons, pp 836-837):

30



$$pK_b = \log_{10} \left(\frac{[\text{RNH}_3^+] [\text{OH}^-]}{[\text{RNH}_2]} \right) \quad (2)$$

5

where R is an aromatic group containing organic structure.

The aromatic amine containing acidic stripping solution is subsequently neutralised to alkaline pH and the aromatic amines return to undissociated form and precipitate out of solution as organic liquids or solids. The organic liquids or solids are separated from the acidic stripping solution. The separated acidic stripping solution may contain saturation levels of undissociated aromatic amines and may be cycled back to the aqueous fluid to undergo further stripping. In the present invention the extraction and acidic stripping solution regeneration stages are integrated so that the streams leaving the process are pure organic liquid and treated aqueous waste respectively.

Preferably the aqueous fluid is an aqueous process stream.

Preferably the aqueous fluid is contacted with one side of the membrane and wherein the acidic stripping solution is contacted with the other side of the membrane. In a more preferred aspect prior to adjusting the pH of the aromatic amine containing acidic stripping solution, the acidic stripping solution is removed from contact with the membrane.

Preferably the acidic stripping solution separated in step (e) is recycled to the aqueous fluid prior to contact with the membrane. In one preferred alternative the acidic stripping solution separated in step (e) is recycled to the aromatic amine containing acidic stripping solution prior to removing the acidic stripping solution from contact with the membrane.

The resulting aromatic amine of step (e) may be a liquid or a solid.

The membrane of the present invention can be configured in accordance with any of the designs known to those skilled in the art, such as spiral wound, plate and frame, shell and tube, and derivative designs thereof. The membranes may be of cylindrical or planar
5 geometry.

For shell and tube designs, the membrane comprises one or more tubular membranes each of which has an internal surface which defines an internal volume. In this aspect either the aqueous fluid or the acidic stripping solution is held within the internal volume of the

10 tubular membrane(s) and the other of the aqueous fluid or the acidic stripping solution is in contact with the external surface of the tubular membrane(s). For spiral wound designs, either the aqueous fluid or the acidic stripping solution is within the membrane leaves and the other of the aqueous fluid or the acidic stripping solution is in contact with the external surface of the membrane leaves.

15

It will be appreciated that in an industrial setting preferably the aqueous fluid is held within the internal volume of the tubular membrane(s) and the acidic stripping solution is in contact with the external surface of the tubular membrane(s), and wherein the tubular membrane(s) and the acidic stripping solution are operably contained.

20

The membrane of the present invention is formed from or comprises a material suitable to provide a non-porous, selectively permeable membrane. The membrane may consist of a homogeneous membrane such as a tube or sheet of material, or a composite membrane. The composite membrane may comprise a non-porous, selectively permeable layer and
25 one or more further materials or may comprise a mixture of materials. The non-porous, selectively permeable layer or material prevents direct contact of the aqueous stream with the acidic stripping solution. This is important. If a direct contact stripping device such as a packed or plate column or microporous membrane contactor is used, the two streams would mix and there would be no resulting separation.

30

In a preferred aspect the membrane or the non-porous, selectively permeable layer thereof

is formed from or comprises a material selected from modified polysiloxane based elastomers including polydimethylsiloxane (PDMS) based elastomers, ethylene-propylene diene (EPDM) based elastomers, polynorbornene based elastomers, polyoctenamer based elastomers, polyurethane based elastomers, butadiene and nitrile butadiene rubber based elastomers, natural rubber, butyl rubber based elastomers, polychloroprene (Neoprene) based elastomers, epichlorohydrin elastomers, polyacrylate elastomers, polyethylene, polypropylene, polytetrafluoroethylene (PTFE), polyvinylidene difluoride (PVDF) based elastomers, and mixtures thereof.

10 In a preferred aspect the membrane comprises a reinforcing material selected from an external mesh and support. This is particularly advantageous for homogenous tubes or sheets. Such tubes or sheets may be reinforced to increase their burst pressure, for example by overbraiding tubes using fibres of metal or plastic, or by providing a supporting mesh for flat sheets.

15 When the membrane comprises a non-porous layer and an additional component, the additional component may be a supporting layer. The supporting layer may be a porous support layer. Suitable materials for the open porous support structure are well known to those skilled in the art of membrane processing. Preferably the porous support is formed
20 from or comprises a material selected from polymeric material suitable for fabricating microfiltration, ultrafiltration, nanofiltration or reverse osmosis membranes, including polyethylene, polypropylene, polytetrafluoroethylene (PTFE), polyvinylidene difluoride (PVDF) polyethersulfone, and mixtures thereof.

25 Preferably the tubular membranes have a high length to diameter ratios for example the tubular membranes may have an internal diameters from 0.5 to 5.0 mm, and/or a wall thicknesses between 0.1 and 1.0 mm and/or a length of from 50 to 500 metres. The length to diameter ratio of the tubular membrane may be from 1×10^4 to 1×10^6 .

30 High length to diameter ratio such as those given above are considerably longer than the length to diameter ratios of membranes typically applied in prior art membrane extraction

processes, and have the advantage that the aqueous fluid entering the membrane tubes passes down a long flow path before emerging from the membrane. Thus it is possible to remove a high percentage of the aromatic amine contaminants in one pass down a single membrane tube, and this reduces the need for extensive manifolding which arises when 5 the aqueous fluid must be passed through several or many membrane modules to achieve the desired degree of removal. This reduction in manifolding results in cost advantages over shorter membrane tubes.

In a further preferred aspect of the present invention a pH control system is used to 10 regulate the flow of acidic stripping solution which contacts the membrane.

Control of pH in the acidic stripping solution is important. Upon contact with the membrane the acidic stripping solution pH will tend to be increased by the dissociation of the aromatic amine, and it is advantageous for the process efficiency that the pH of the 15 acidic stripping solution is kept at least 0.5 pH units below the pKa of the aromatic amine. This may be achieved by fixing the flowrate and strength of the acidic stripping solution so as to ensure that this condition is always met. A higher acid concentration in the acidic stripping solution for given volumes or flows of aqueous fluid and acidic stripping solution will meet this condition better than a lower concentration of acid. A higher acid 20 concentration also makes possible a lower acid flowrate for a given aromatic amine loading in the aqueous fluid; this results in a lower recycle stream flowrate from step (e), and hence a more cost effective system. However use of excessive acid in the acidic stripping solution will require excess alkali in the recovery stage. Also, excessively strong acids may damage the non-porous membrane. For example, silicone rubber is one 25 of the most permeable elastomers and so desirable for use in the process, but can be damaged by acid concentrations above 1 wt.% mineral acid.

Preferably the stripping solution in contact with the nonporous membrane is well mixed so that its composition is well mixed throughout the volume operably in contact with the 30 nonporous membrane.

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Preferably the pH of the acidic stripping solution in contact with the non-porous membrane is controlled so that it is substantially the same throughout the acidic stripping solution in contact with the non-porous membrane separating layer.

5 Preferably the aqueous fluid contains an aromatic amine selected from aniline, chloroanilines, dichloroanilines, toluidines, dimethylaniline, nitroaniline, phenylaniline, methylaniline, ethylaniline, anisidine, benzylamine, fluoroanilines, difluoroanilines, aminophenols, and mixtures thereof.

10 Preferably the acidic stripping solution comprises a mineral acid selected from hydrochloric acid, sulphuric acid, phosphoric acid, nitric acid, and mixtures thereof.

Preferably the pH of the aromatic amine containing acidic stripping solution is adjusted in step (d) by the addition of an alkali.

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Preferably the alkali is an aqueous solution of an alkali selected from sodium hydroxide, sodium carbonate, calcium hydroxide, calcium carbonate, potassium hydroxide, potassium carbonate, and mixtures thereof.

20 In a further preferred aspect the aqueous fluid is contacted with one side of a plurality of membranes in series, in parallel or in a combination thereof, and wherein the acidic stripping solution is contacted with the other side of each of the plurality of membranes.

25 The process may be performed in a continuous, semi-continuous or discontinuous (batch mode) manner. In the latter aspect the flow of at least one of the aqueous fluid, the acidic stripping solution, and the alkali solution is discontinuous.

In one aspect the resulting aromatic amine of step (e) is contacted with an organic solvent and subsequently treated in a further process. In this aspect it may be desirable to contact
30 the amine containing acidic stripping solution and/or the separated aromatic amine with a solvent or solvent mixture in step (e). This may be particularly useful when the separated

aromatic amine is a solid. The solvent introduced may dissolve the solid. This may be further useful when this solid is a product or reactant in a reaction and where the solid and the solvent used to dissolve the solid can be sent to the further process in which the solid material is produced or consumed.

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The process of the present invention may be performed in a reactor comprising at least a first zone, a second zone, a third zone, and a fourth zone; wherein each of the zones is discrete from each other zone; wherein the first zone and the second zone are separated by the non porous membrane; wherein the first zone contains the aqueous fluid ; wherein the

10 second zone and fourth zone contain the acidic stripping solution; wherein the third zone contains amine containing acidic stripping solution; wherein the first zone and the third zone are operably connected to each other; wherein the second zone is operably connected to the third zone and the fourth zone; and wherein the acidic stripping solution is circulated between the fourth zone and the second zone such that the acidic stripping solution is well mixed throughout its volume.

15 Preferably, the acidic stripping solution is circulated between the fourth zone and the second zone at a high rate relative to the flow of aqueous fluid. By the term "high rate" it is preferably meant that the volume of acidic stripping solution contacted with the
20 membrane is greater than the volume of aqueous fluid contacted with the membrane. The ratio of acidic stripping solution volume to aqueous fluid volume contacted with the membrane may be >2:1, >5:1, or >10:1.

25 The aqueous fluid and/or the acidic stripping solution of the present invention may be heated before contact with the membrane. The aqueous fluid and/or the acidic stripping solution of the present invention may have a temperature above room temperature (25°C). This may increase the rate of mass transfer across the non-porous membrane.

30 In a further preferred aspect the aqueous fluid contains substantial quantities of dissolved inorganic or organic materials. By the term "substantial quantities" it is meant greater than 0.1 wt%. The inorganic materials may include salts, such as sodium chloride,

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potassium chloride and mixtures thereof. The organic materials may include solvents, such as methanol, ethanol, acetone, acetate and mixtures thereof.

The aromatic amine in the acidic stripping solution dissociates according to an equilibrium reaction described by equation (1). Even at low pH, there will be some finite fraction of the aromatic amine present in dissociated form, and the total aromatic amine concentration will be equal to the sum of the concentration of dissociated and the concentration of undissociated aromatic amine. In general, the higher the concentration of total aromatic amine in the acidic stripping solution at a given pH, the higher will be the concentration of undissociated aromatic amine. This undissociated aromatic amine will act to reduce the driving force for mass transfer of undissociated aromatic amine from the aqueous fluid to the acidic stripping solution. This effect will be relatively greater for the aqueous fluid in the section of membrane near the point of exit of the aqueous fluid from the membrane.

15

Thus in a further preferred embodiment of the present invention, it is desirable to use two well mixed stripping stages in series. In this embodiment, the aqueous fluid first contacts a membrane whose other side is in contact with a well mixed strength 1 acidic stripping solution in a first stripping stage, and then contacts a second membrane whose other side is in contact with a well mixed strength 2 acidic stripping solution in a second stage. Strength of an acidic stripping solution is determined by the strength of the acid, for example, the mineral acid, fed to the acidic stripping solution. In this aspect, the mineral acid concentration fed to stripping solution 1 is stronger than the mineral acid concentration fed to stripping solution 2. The aqueous fluid passes from the membrane of stripping stage 1 to membrane of stripping stage 2. Mineral acid is fed to the acidic stripping solution in stripping stage 2, and the resulting strength 2 stripping solution from stage 2 is passed into stage 1 where further mineral acid is added to increase the strength of the acidic stripping solution in stage 1 to strength 1. The total aromatic amine concentration in stage 1 is greater than the total aromatic amine concentration in stage 2. The pH may be controlled to be constant in each stripping stage and may be set at different values in stage 1 and stage 2. The use of more than two stages is also envisaged.

The invention will now be described, by way of example only, with reference to the accompanying drawings, in which:-

- 5 Figure 1 is a schematic of an apparatus operating the process of the present invention.
- Figure 2 is a schematic of an apparatus operating the process of the present invention.
- Figure 3 is a schematic of an apparatus operating the process of the present invention.
- Figure 4 is a schematic of an apparatus operating the process of the present invention.
- Figure 5 is a schematic of an apparatus operating the process of the present invention.
- 10 Figure 6 is a schematic of an apparatus operating the process of the present invention.

Figure 1 shows a schematic of one embodiment of the process. The aqueous fluid containing undissociated aromatic amines (1) passes on one surface of a membrane containing at least one non-porous separating layer (5), optionally mounted in a membrane module (6). Undissociated aromatic amines in the wastewater permeate across the membrane into the acidic stripping solution (3), whose pH is such that the aromatic amines are converted into their corresponding salts. The aqueous fluid exiting the membrane has a reduced concentration of aromatic amines relative to the aqueous fluid (1) entering the membrane. The aromatic amine laden acidic stripping solution (4) leaves the membrane module (6) containing dissociated aromatic amines and enters a neutralisation vessel (7). By manipulation of the ratio between the volume of aqueous stream fed (1) and volume of acidic stripping solution fed (3), i.e. by using a ratio greater than 1, the concentration of dissociated aromatic amines in aromatic amine laden acidic stripping solution (4) is elevated to levels higher than the saturation concentration of undissociated aromatic amines in water. In the neutralisation vessel (7) a mineral alkali (8) is added to adjust pH of the solution to a value above the pKa of the aromatic amine. This converts the aromatic amine back to an undissociated form. Since it is at a concentration higher than the saturation concentration of undissociated amine in water, the aromatic amine precipitates out of solution as a liquid or solid. The neutralisation vessel (7) may be optionally stirred. The resulting two phase mixture (9) is passed to a settling vessel (10) where the two phases are separated. The organic phase (either liquid

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or solid) is removed (11) from the settling vessel (10), and the aromatic amine saturated aqueous layer (12) is recirculated back either to the aqueous process stream (1), or to the neutralisation tank (7).

5 In a preferred embodiment, the membranes may comprise a bundle of tubular membranes with suitable head piece connections for allowing flow of the aqueous fluid to pass through the interior of the membranes. This bundle of tubular membranes may be suspended in a tank or other vessel so that the outside surface of the fibres is substantially immersed in the acidic stripping solution. In this case the acidic stripping solution will be
10 mixed or agitated using a stirrer or pump or some other suitable device to ensure that the acidic stripping solution is well mixed at all times and the composition of the stripping solution in contact with the membrane will be the same as the concentration of the stripping solution (4) leaving the tank (15). Figure 2 shows this general arrangement where a bundle of tubular membranes (13) are connected at each end to allow wastewater
15 flow through headpieces (14), and are immersed in a tank (15) of acidic stripping solution.

Figure 3 shows yet another preferred embodiment, in which one or more elastomeric tubular membranes (16) connected using suitable headpieces (17) are suspended in a well mixed tank (15) containing acidic stripping solution (4). The elastomeric tubular
20 membranes can be coiled, stacked or otherwise arranged in the tank so that they have their surfaces substantially immersed in the acidic stripping solution (4). It is advantageous in this embodiment to use elastomeric tubular membranes which have high length to diameter ratios for example the elastomeric tubular membranes might have internal diameters from 0.5 to 5.0 mm, wall thicknesses between 0.1 and 1.0 mm and lengths from
25 50 to 500 metres, i.e. length to diameter ratios of 1×10^4 to 1×10^6 .

The configurations illustrated in Figures 2 and 3 are made possible by the rapid dissociation reaction in the acidic stripping solution which removes the need to configure the flow over the outside surfaces of the membrane so as to provide high rates of film
30 mass transfer.

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It is apparent to one skilled in the art that it is desirable to use a strong acid (by way of non limiting example, hydrochloric acid above 20 wt.% (HCl), as a feed acidic stripping solution (3), while maintaining pH in the acidic stripping solution in contact with the non-porous membrane (4) separating layer at a high enough value to minimise the need for alkali addition and to ensure maximal lifetime of the non-porous membrane separating layer. It is with these objectives in mind that the configurations of Figures 2, 3, 4 and 5 are found to have advantages over passing the aqueous and organic streams in countercurrent flow through membrane modules as shown in Figure 1. In the configurations of Figures 2, 3, 4 and 5 the acid added (3) can be concentrated but as it is immediately mixed into the aromatic amine laden acidic stripping solution (4) in the well mixed tank (15) the actual acid concentration of the acidic stripping solution in contact with the non-porous membrane separating layers can be everywhere substantially less than the acid concentration of the feed acidic stripping solution (3).

[+/- 10]
[+/- 11]
[+/- 12]
[+/- 13]
[+/- 14]
[+/- 15]
[+/- 16]
[+/- 17]
[+/- 18]
[+/- 19]
[+/- 20]

15 A further preferred embodiment of the process as shown in Figure 4 by way of non-limiting example can be employed. A well mixed tank (15) containing an elastomeric membrane tube (16) immersed in acidic stripping solution (4) has a pH sensor (18) in contact with the well mixed acidic stripping solution (4). This pH sensor measures pH and transmits this information to a control device (19) which regulates flow of feed acidic
20 stripping solution (3) to the tank to hold pH at the desired value. Using this approach pH in the tank (15) can be controlled to the highest value consistent with good process efficiency, thus minimising acidity in the tank and in the aromatic amine laden acidic stripping solution (4). This has consequent advantages for neutralisation costs and membrane lifetime.

25

In yet another preferred embodiment, shown in Figure 5, the advantages of a well mixed acidic stripping solution in a remote stripping tank (20) are shown in connection with the use of a membrane module of the type used in Figure 1. The pH is controlled in the stripping tank by a pH sensor (18) and a control device (19) which regulates the flow feed
30 acidic stripping solution (3) to the stripping tank (20). The acidic stripping solution is recirculated (21) to the membrane module or modules at a high rate so that for all

practical purposes the acidic stripping solution can be considered well mixed throughout its volume. Aromatic amine laden acidic stripping solution is withdrawn (4) and passed to the neutralisation tank (7).

- 5 In yet another preferred embodiment the temperature of the acidic stripping solution in tank (15) or tank (20) and/or the aqueous stream (1) can be increased above ambient conditions to increase the rate of mass transfer across the non-porous separating layer of the membrane.
- 10 In yet another preferred embodiment shown in Figure 6, the use of two stripping stages is shown with two strengths of acidic stripping solution. The aqueous fluid (1) enters the first stripping stage in which a membrane (30) is suspended in the first stage tank (31). An acidic stripping solution containing mineral acid (32) is added to the stage 1 tank (31) automatically by a pH controller (34) connected to a pH probe (33). The stripping solution in stage 1 is well mixed throughout the volume of the stage 1 tank (31) so that it is of identical composition to the stripping solution (42) exiting the stage 1 tank. The aqueous fluid (35) flows out of stage 1 and into stage 2 where a second membrane (40) is suspended in the stage 2 tank (41). The aqueous fluid (2) flows out of the second membrane (40) with a reduced concentration of aromatic amine relative to stream (1). In
- 15 20 the second stage, acidic stripping solution containing mineral acid (37) at a lower concentration than stream (32) is added to the stage 2 tank (41) automatically by a pH controller (39) connected to a pH probe (38). The stripping solution in the stage 2 tank (41) is well mixed throughout the volume of the stage 2 tank so that it is of identical composition to the stripping solution (36) exiting the stage 2 tank. The stripping solution
- 25 30 exiting the stage 2 tank (41) is fed to the stage 1 tank (31). The system is configured and operated so that the total aromatic amine concentration in stream (36) is less than the total aromatic amine concentration in stream (42). The stripping solution (42) from the stage (1) tank is passed to the neutralisation vessel (7). Following phase separation, the aromatic amine saturated aqueous layer (12) is recirculated back to the aqueous fluid (1).

batch mode. The tanks may be single tanks or multiples tanks. The neutralisation vessel (7) and the phase separating vessel (10) may be combined into the same vessel. Mixing of one or all of the tanks may be achieved by using any device known to those skilled in the art, such as mixers, pumps, or air lift devices. Variations and changes may be made by 5 those skilled in the art without departing from the spirit of the invention.

The invention will now be described in further detail in the following non-limiting Examples.

10 EXAMPLES

EXAMPLE 1

15 The following example describes batch operation of the present invention. 1000 mL of acidic stripping solution comprising a dilute solution of hydrochloric acid were added to a stirred tank containing a 13 metre length of a silicone rubber membrane tube with internal diameter of 3 mm, wall thickness 0.6 mm. 20 litres of a wastewater containing 0.6 wt.% aniline were recirculated from a drum through the inside of the elastomeric membrane tube. Concentrated (37 wt.%) hydrochloric acid was added regularly to the acidic 20 stripping solution to maintain pH in the range 1-4. After 14 days the experiment was stopped, and the acidic stripping solution removed. The pH of the acidic stripping solution was adjusted to 11 by addition of 40 wt.% NaOH solution, and a dark organic liquid formed and was separated. This organic liquid was analysed via gas chromatography and found to be more than 99% pure aniline.

25

EXAMPLE 2

30 The following example describes continuous operation of the present invention. 1000 mL of acidic stripping solution were added to a stirred tank containing a 10 metre length of a silicone rubber membrane tube with internal diameter of 3 mm, wall thickness 0.6 mm. A pH sensor was suspended in the tank and a controller was connected to the sensor so as to

add 37 wt.% hydrochloric acid to the tank when required to maintain pH. The controller held pH at 2.5 +/- 0.2 pH units. A flow of aqueous process stream containing aniline at 0.5 wt.% was pumped from a drum and passed through the inside of the membrane tube. The overflow of the acidic stripping solution was periodically removed to a tank where 5 pH was adjusted, resulting in formation of a dark organic liquid. The resulting aqueous and organic phases were separated and the aqueous phase mixed with the aqueous process stream in the drum. The dark organic liquid was analysed and found to be aniline.

EXAMPLE 3

10

The following example describes batch operation of the present invention used to recover 2,4 difluoroaniline from an aqueous stream. 1000mL of strip solution comprising a dilute solution of hydrochloric acid were added to a stirred tank containing a 50 metre length of a silicone rubber membrane tube with internal diameter of 3mm, wall thickness 0.6 mm.

15

20 litres of a wastewater containing 0.35wt% 2,4 difluoroaniline were recirculated from a drum through the inside of the elastomeric membrane tube. Concentrated (37wt%) hydrochloric acid was added regularly to the strip solution to maintain pH in the range 1.5-2.0. After 4 days the experiment was stopped, and the strip solution removed. The pH of the strip solution was adjusted to 11 by addition of 40wt% NaOH solution, and a dark 20 organic liquid formed and was separated. This organic liquid was analysed via gas chromatography and found to be more than 99% pure 2,4 difluoroaniline.

EXAMPLE 4

25

The following example describes batch operation of the process to recover a mixture of 2,4 difluoroaniline and 2,4,6 bromodifluoroaniline from an aqueous waste. 1000mL of strip solution comprising a 10%wt solution of hydrochloric acid were added to a stirred tank containing a 20 metre length of an EPDM rubber membrane tube with internal diameter of 3mm, wall thickness 1 mm. 20 litres of a wastewater containing 0.35wt% 2,4 30 difluoroaniline and 0.15 wt% 2,4,6 bromodifluoroaniline were recirculated from a drum through the inside of the elastomeric membrane tube. After 4 days the experiment was

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stopped, and the strip solution removed. The pH of the strip solution was adjusted to 11 by addition of 40wt% NaOH solution, and a dark organic liquid formed and was separated. This organic liquid was analysed via gas chromatography and found to be more than 20% 2,4 difluoroaniline and 80% 2,4,6 bromodifluoroaniline.

5

All publications mentioned in the above specification are herein incorporated by reference. Various modifications and variations of the described methods and system of the invention will be apparent to those skilled in the art without departing from the scope

10 and spirit of the invention. Although the invention has been described in connection with specific preferred embodiments, it should be understood that the invention as claimed should not be unduly limited to such specific embodiments. Indeed, various modifications of the described modes for carrying out the invention which are obvious to those skilled in chemistry or related fields are intended to be within the scope of the
15 following claims.

CLAIMS

1. A process for removing and recovering one or more undissociated aromatic amines dissolved in aqueous fluid, the process comprising the steps of:
 - 5 (a) transferring the one or more undissociated aromatic amines from the aqueous fluid to an acidic stripping solution, wherein transfer of the one or more undissociated aromatic amines from the aqueous fluid to the acidic stripping solution occurs across a membrane; wherein the membrane is a non porous, selectively permeable membrane;
 - (b) regulating the volume of acidic stripping solution employed relative to the volume of aqueous fluid treated so that the total aromatic amine concentration in the acidic stripping solution, comprising the sum of the dissociated and undissociated aromatic amine concentrations, is above the solubility of the aromatic amines in water;
 - 10 (c) regulating the pH of the acidic stripping solution in contact with the membrane so that the membrane remains selectively permeable;
 - (d) adjusting the pH of the aromatic amine containing acidic stripping solution to a value above the acidic dissociation constant of the aromatic amine and
 - (e) separating the resulting aromatic amine and the acidic stripping solution.
2. A process according to claim 1 wherein the aqueous fluid is an aqueous process stream.
- 20 3. A process according to claim 1 or 2 wherein the aqueous fluid is contacted with one side of the membrane and wherein the acidic stripping solution is contacted with the other side of the membrane.
- 25 4. A process according to claim 3 wherein prior to adjusting the pH of the aromatic amine containing acidic stripping solution, the acidic stripping solution is removed from contact with the membrane.
- 30 5. A process according to claim 3 or 4 wherein the acidic stripping solution separated in step (e) is recycled to the aqueous fluid prior to contact with the membrane.

6. A process according to claim 4 wherein the acidic stripping solution separated in step (e) is recycled to the aromatic amine containing acidic stripping solution prior to removing the acidic stripping solution from contact with the membrane.

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7. A process according to any one of the preceding claims wherein the resulting aromatic amine of step (e) is a liquid.

8. A process according to any one of claims 1 to 6 wherein the resulting aromatic amine of step (e) is a solid.

10

9. A process according to any one of the preceding claims wherein the membrane is mounted in a plate and frame configuration, a shell and tube configuration, or a spiral wound configuration.

15

10. A process according to any one of the preceding claims wherein the membrane comprises one or more tubular membranes each of which has an internal surface which defines an internal volume and either the aqueous fluid or the acidic stripping solution is held within the internal volume of the tubular membrane(s) and the other of the aqueous fluid or the acidic stripping solution is in contact with the external surface of the tubular membrane(s).

20

11. A process according to claim 10 wherein the aqueous fluid is held within the internal volume of the tubular membrane(s) and the acidic stripping solution is in contact with the external surface of the tubular membrane(s), and wherein the tubular membrane(s) and the acidic stripping solution are operably contained.

25

12. A process according to claim 10 or 11 wherein the tubular membrane(s) is elastomeric.

30

13. A process according to any one of claims 10 to 12 wherein the tubular

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membrane(s) has a length to diameter ratio of from 10.000 to 1.000.000.

14. A process according to any one of the preceding claims wherein the acidic stripping solution in contact with the nonporous membrane is well mixed throughout its

5 volume such that its composition is uniform throughout.

15. A process according to any one of the preceding claims wherein the pH of the acidic stripping solution in contact with the non-porous membrane is controlled so that it is substantially the same throughout the volume of acidic stripping solution in contact
with the non-porous membrane separating layer.

16. A process according to any one of the preceding claims wherein the aqueous fluid contains an aromatic amine selected from aniline, chloroanilines, dichloroanilines, toluidines, dimethylaniline, nitroaniline, phenylaniline, methylaniline, ethylaniline, anisidine, benzylamine, fluoroanilines, difluoroanilines, aminophenols and mixtures thereof.

17. A process according to any one of the preceding claims wherein the acidic stripping solution comprises a mineral acid selected from hydrochloric acid, sulphuric acid, phosphoric acid, nitric acid, and mixtures thereof.

18. A process according to any one of the preceding claims wherein the pH of the aromatic amine containing acidic stripping solution is adjusted in step (d) by the addition of an alkali.

25

19. A process according to claim 18 wherein the alkali is an aqueous solution of an alkali selected from sodium hydroxide, sodium carbonate, calcium hydroxide, calcium carbonate, potassium hydroxide, potassium carbonate, and mixtures thereof.

30

20. A process according to any one of the preceding claims wherein the membrane is formed from or comprises a material selected from modified polysiloxane based

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elastomers. ethylene-propylene diene (EPDM) based elastomers. polynorbornene based elastomers. polyoctenamer based elastomers. polyurethane based elastomers. butadiene and nitrile butadiene rubber based elastomers. natural rubber. butyl rubber based elastomers. polychloroprene (Neoprene) based elastomers. epichlorohydrin elastomers. 5 polyacrylate elastomers. polyethylene. polypropylene. polytetrafluoroethylene (PTFE). polyvinylidene difluoride (PVDF) based elastomers, and mixtures thereof.

21. A process according to claim 20 wherein the modified polysiloxane based elastomer is a polydimethylsiloxane (PDMS) based elastomer.

10

22. A process according to any one of the preceding claims wherein the membrane comprises a reinforcing material selected from an external mesh and support.

15

23. A process according to any one of the preceding claims wherein the membrane is a composite membrane comprising a porous support and at least one non-porous layer.

20

24. A process according to claim 23 where the porous support is formed from or comprises a material selected from polymeric material suitable for fabricating microfiltration. ultrafiltration. nanofiltration or reverse osmosis membranes, including polyethylene. polypropylene. polytetrafluoroethylene (PTFE), polyvinylidene difluoride (PVDF) polyethersulfone, and mixtures thereof.

25. A process according to any one of the preceding claims wherein the aqueous fluid is contacted with one side of a plurality of membranes in series. in parallel or in a combination thereof, and wherein the acidic stripping solution is contacted with the other side of each of the plurality of membranes.

26. A process according to any one of the preceding claims wherein the process is performed in a continuous manner.

30

27. A process according to any one of claims 19 to 25 wherein the flow of at least one

of the aqueous fluid, the acidic stripping solution, and the alkali solution is discontinuous.

28. A process according to any one of the preceding claims wherein the resulting aromatic amine of step (e) is contacted with an organic solvent and subsequently treated
5 in a further process.

29. A process according to any one of the preceding claims wherein the process is performed in a reactor comprising at least a first zone, a second zone, a third zone, and a fourth zone

10 wherein each of the zones is discrete from each other zone;
wherein the first zone and the second zone are separated by the non porous membrane;
wherein the first zone contains the aqueous fluid ;
wherein the second zone and fourth zone contain the acidic stripping solution;
wherein the third zone contains amine containing acidic stripping solution;
15 wherein the first zone and the third zone are operably connected to each other;
wherein the second zone is operably connected to the third zone and the fourth zone;
and wherein the acidic stripping solution is circulated between the fourth zone and the second zone such that the acidic stripping solution is well mixed throughout its volume.

20 30. A process according to claim 29 wherein a pH control system is used to regulate the flow of acidic stripping solution between the fourth zone and the second zone.

31. A process according to any one of the preceding claims wherein at least one of the aqueous fluid and the acidic stripping solution has a temperature above room temperature.

25 32. A process according to any one of the preceding claims wherein the aqueous fluid contains substantial quantities of dissolved inorganic or organic materials.

33. A process as substantially described herein and with reference to any one of
30 Figures 1-6.

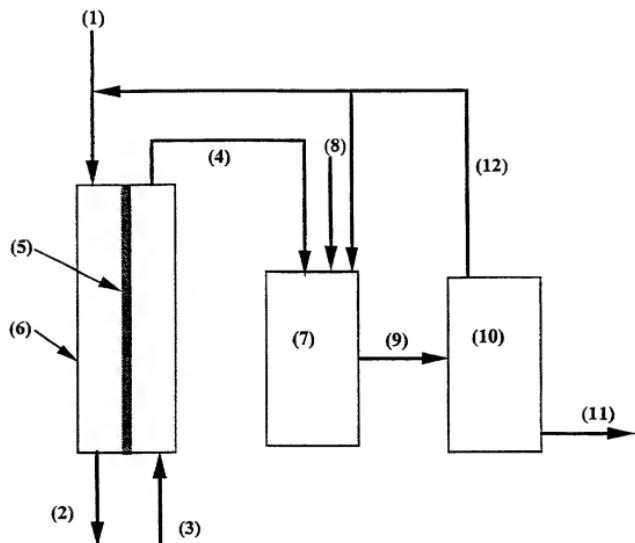


Figure 1 - Aromatic amine recovery process

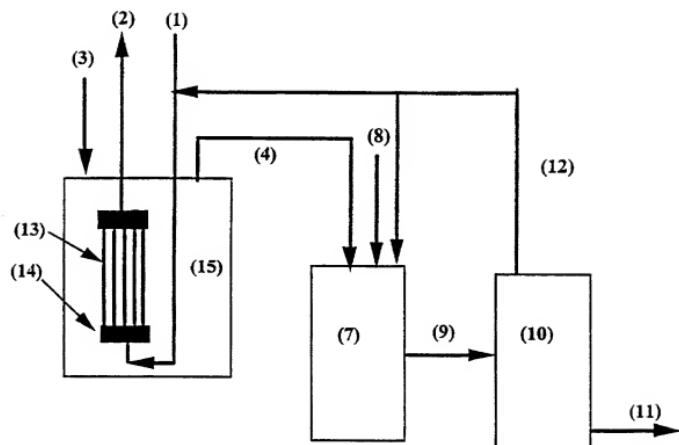


Figure 2- Aromatic amine recovery process with
bundle of tubular membranes

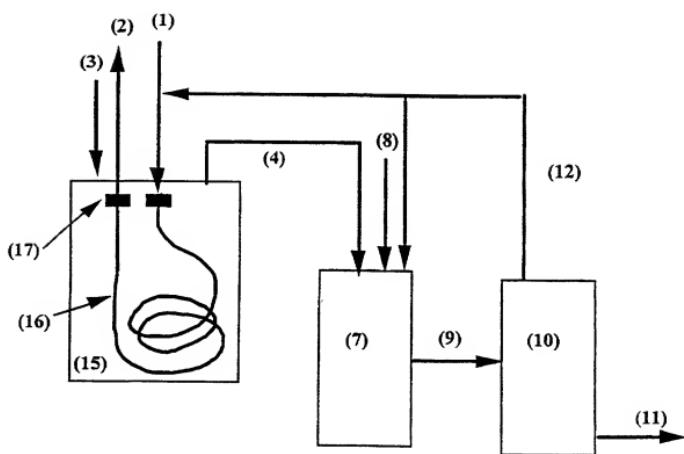


Figure 3- Aromatic amine recovery process with elastomeric tube membrane

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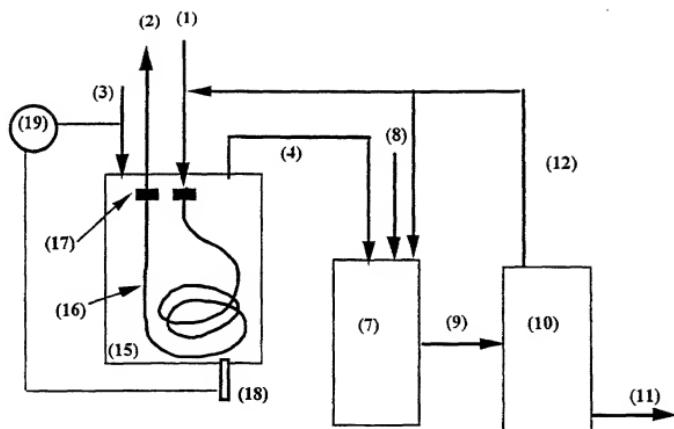


Figure 4- aromatic amine recovery process with
elastomeric tube membrane and pH control

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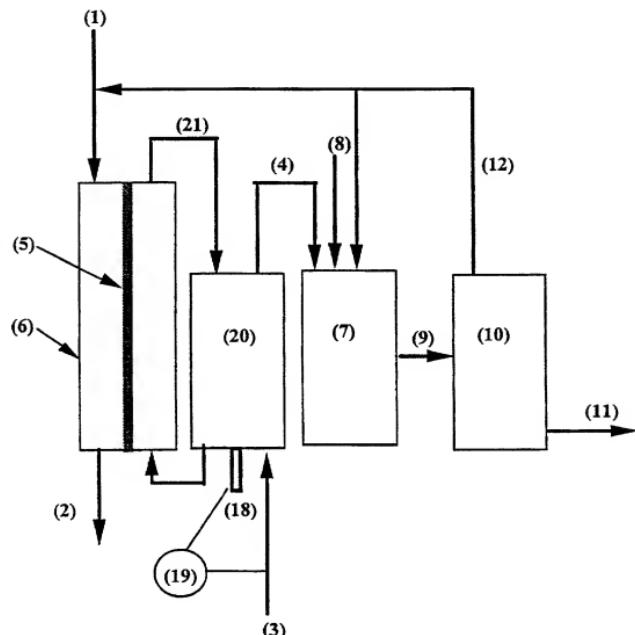


Figure 5 - Aromatic amine recovery process operated with a well mixed stripping solution recirculated at high rate to a remote membrane module.

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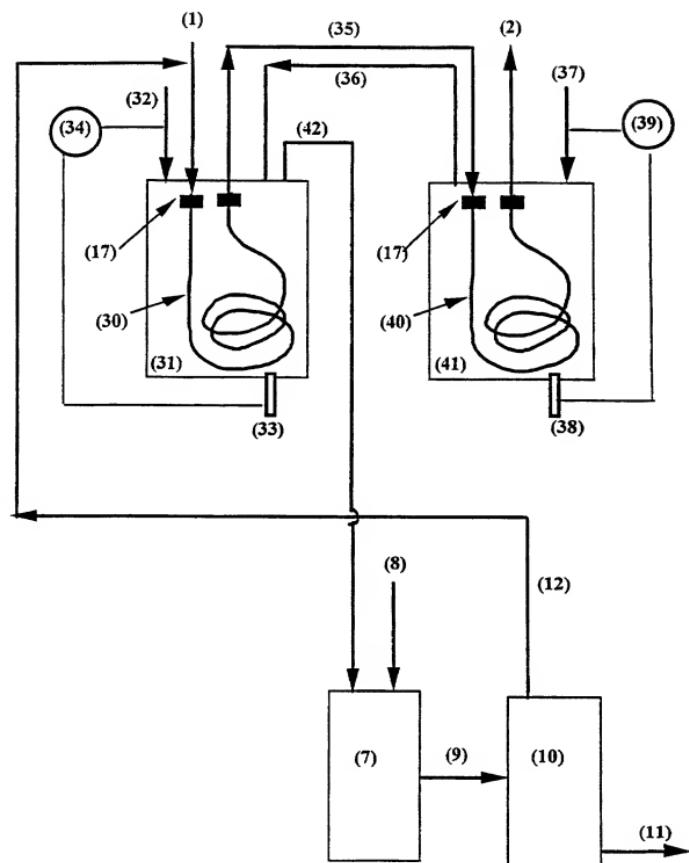


Figure 6 - Two stage aromatic amine recovery process

Attorney Docket No YOUN 2 0076

DECLARATION AND POWER OF ATTORNEY FOR PATENT APPLICATION

As a below Inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor(s) if only one name is listed below or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

**METHOD FOR REMOVING AND RECOVERING
AROMATIC AMINES BY USING A NON-POROUS MEMBRANE**

the specification of which
 is attached hereto

[] was filed on _____
 Application Serial No. _____
 and was amended on _____
 (if applicable)

[] was filed _____

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, § 1.56 (a).

I hereby claim foreign priority benefits under Title 35, United States Code, § 119 or 365(b) of any foreign application(s) for patent or inventor's certificate, or 365(a) of any PCT international application which designated at least one country other than the United States of America, listed above and have also identified below, by check the box, any foreign application(s) for patent or inventor's certificate, or of any PCT international application having a filing date before that of the application on which priority is claimed:

Prior Foreign Application

9918U31.7	<input checked="" type="checkbox"/>	Great Britain	30/July/1999	No
(Number)	(Country)	(Day/Month/Year Filed)	Certified Copy Attached?	

I hereby claim the benefit under 35 U.S.C. 119(e) of any United States provisional application(s) listed below.

Application No(s)	<input checked="" type="checkbox"/>	J additional provisional application numbers are listed on a supplemental priority data sheet attached
	(Day/Month/Year Filed)	

I hereby claim the benefit under Title 35, United States, § 120 of any United States application(s) or any PCT international application designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose material information which is material to patentability as defined in Title 37, of Federal Regulations Code, § 1.56(a) which became available between the filing date of the prior application and the national or PCT international filing date of this application:

PCT/GB00/02827	<input checked="" type="checkbox"/>	July 21, 2000	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (If Applicable)
U.S. Patent Application or PCT Parent Number				

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to

Attorney Docket No. YOU 2 0076

prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

30-

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both under § 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

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